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Radiological Health

NATURAL ENVIRONMENTAL RADIOACTIVITY FROM RADON 222

U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE

Public Health Service

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by

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ABSTRACT

Radium 226 and radon 222 have been measured using a deemanation method in which the gaseous daughter, radon 222, is collected and analyzed by an alpha-scintillation method. "Emanating radium 226" from particles of soil in clay type soils varied from 0.342×10^{-12} grams per gram of dry soil to 0.690×10^{-12} grams per gram. Sandy soils varied between 0.147×10^{-12} to 0.244×10^{-12}. Highest activity measured was 14.3×10^{-12} grams of "emanating radium 226" per gram of dry soil containing uranium ore.

Emanation from ground in Champaign County, Ill., for 216 measurements, was $(140 \pm 73) \times 10^{-18}$ curies of radon 222 cm^{-2} sec^{-1}. Other soils released similar values of radon 222 except at locations where uranium 238 is available in commercially signifiant amounts.

Release of radon 222 from the leaves of corn (*Zea mays L.*) about midday and near the growth peak was approximately 2.6 times that of the soil under the corn at Argonne National Laboratory in one series of six tests.

Radon 222 concentrations in the atmosphere were measured in eight series of 24-hour tests, to elevations of 16 m. Values of concentration at various times of the day are presented for the normal environmental levels.

The use in this report of trade names of commercially available products is only to indicate that the particular product is being used in the laboratory. Such use of the name or the product does not constitute an endorsement of the product of the Public Health Service nor the U.S. Department of Health, Education, and Welfare, and does not imply that there are no other suitable products available commercially.

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NATURAL ENVIRONMENTAL RADIOACTIVITY FROM RADON 222

INTRODUCTION

Early in this century Elster and Geitel (1901) (*1*) reported that the atmosphere contained radioactive substances. Rutherford and Allen (1902) (*2*) observed that the radioactivity of the natural atmosphere resembled that of radon and thoron. Since that time, a number of scientists have made contributions to our knowledge of radon, thoron, actinon, and radioactive gases in the natural atmosphere.

Radon (3.8 days half-life) has been more amenable to study and is of greater interest as a health hazard than thoron (54.5 sec) and actinon (3.9 sec) because radon has the longer half-life. Since 1958, studies of radon 222 in the atmosphere have been made at Argonne National Laboratory and at the University of Illinois, Urbana. These studies have included:

1. The amount of radium 226 in soils capable of releasing radon 222 to the interpore space for eventual diffusion toward the surface and the atmosphere.
2. The amount of radon 222 released from the soil surface or from leaves of plants into the atmosphere.
3. The feasibility of using radon 222 as a free natural radioactive tracer material for studies of vertical diffusion in the atmosphere under a variety of meteorological conditions.
4. Atmospheric concentrations of radon 222 at various heights and times and under a variety of meteorological conditions.

The purpose of this report is to summarize the results of the studies for the scientific community concerned with the amount of human exposure to radioactivity attributable to one of the most important of the natural sources, radon 222.

EXPERIMENTAL METHODS

Radon 222 is a radioactive, noble gas in the decay series of uranium 238 (table 1). Daughters of uranium 238, to and including radium 226, being particulate and ionized attach themselves to nearby solid materials and are relatively immobile.

However, radon 222 being gaseous may be released to pore space between particles and may diffuse in the direction of decreasing concentration gradient, in the soil, toward the surface and into the atmosphere.

The procedures required to trace the origins of radon 222 from its parent radium 226 to diffusion into the atmosphere include: (1) measurement of radon 222 in gas samples, (2) measurements of radon released from particles of soil, and (3) collection and measurement of volumes of gas above the surface of soil beds.

Table 1. Uranium 238 (radium) series

Element		Radiation	Half-life
Uranium	^{238}U	α, γ	4.5×10^{9} yr
.	.	.	.
.	.	.	.
.	.	.	.
Radium	^{226}Ra	α, γ	1.6×10^{3} yr
Radon	^{222}Rn	α, γ	3.8 days
Radium A	^{218}Po	α, β	3.05 min
Radium B	^{214}Pb	β, γ	26.8 min
Radium C	^{214}Bi	α, 99.96 percent β, γ	19.7 min
Radium C^1	^{214}Po	99.96 percent α	1.6×10^{-4} sec
Radium D	^{210}Pb	β, γ	22 yr

Radium 226 and Radon 222 Analysis

Quantities of both radium 226 and radon 222 were determined using the emanation method reported by Stehney *et al.* (*3*), Lucas (*4*), and was summarized by Blanchard (*5*).

The emanation method may be described briefly as follows. Air containing a quantity of radon 222 was passed through Dry Ice chilled traps in which the radon was adsorbed on activated coconut charcoal.

The charcoal traps containing the samples were transferred to the laboratory where the radon samples were desorbed by heating the charcoal to 300° C. The traps were flushed several times, aged helium being used as a carrier, and the radon-helium mixture was pumped into evacuated counting bottles (fig. 1). The counting bottles, covered at one end by a quartz glass window, had been lined with a silver-activated zinc sulfide scintillation material. After the transfers, the bottles were sealed and allowed to stand for about 2 hours during which time the short-lived radon decay products established approximate equilibrium with the parent radon 222.

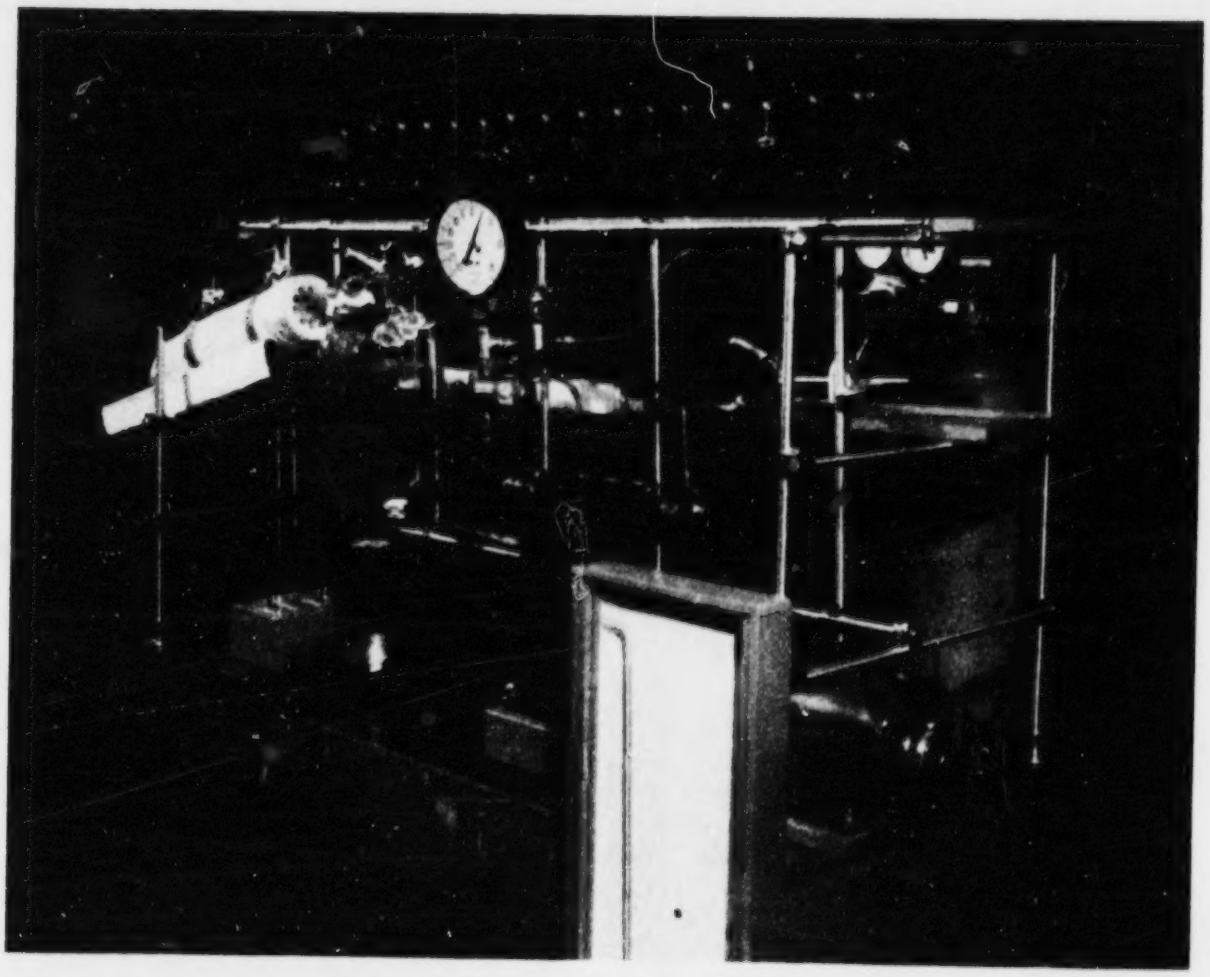

Figure 1. Transfer bench where radon is released from charcoal in the trap and pumped to counting bottles. Counting bottles are stored on upper shelf.

The radon content was determined by placing the counter bottle window in contact with the window of a photomultiplier tube, covering the assembly with a light tight shield, and counting the alpha scintillations for a measured period of time. Corrections for radon decay during the intervals between collection, transfer, and counting were made. For a typical background counting rate of about 0.1 cpm,

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this system has demonstrated an efficiency of 4.85±0.05 (standard deviation) cpm/pCi of radon. The maximum possible cpm/pCi radon 222 is 6.66, because there are three alpha particles per distinegration and 2.22 dpm.

The ability of the charcoal to adsorb radon at greater activity and higher flow rates than normally used was tested by placing two charcoal traps in a series and measuring the activity retained in each trap. The average percentage of activity collected in the first trap was 99.38 percent with a range of 99.20 percent to 99.58 percent for four tests. The flow rate for the four tests was 30 liters/min while the activity ranged from 39 to 266 pCi of radon 222. In the testing program the flow rate was standardized at 4 liters/min with an average activity of 10 pCi of radon 222.

Particle "Emanating Radium 226" Measurements

Particle soil "emanating radium 226" concentrations are defined as the measured quantity of parent radium 226, sufficiently near the surface of soil particles from which the observed quantity of radon 222 would be produced. That portion of radon 222 held within the crystal structure was not of interest although it possesses the capability of providing beta and gamma radiation to the environment.

"Emanating radium 226" of soil particles was determined by placing a carefully weighed sample of about 5 grams of dried soils in a flask with a weak acid solution to simulate normal soil acidity. The flask was then flushed with radon-free air, sealed, and left undisturbed for about 1 week for radon 222 accumulation from its parent, radium 226. After standing, the flask was again flushed and the accumulated radon was transferred to charcoal and the analysis accomplished as previously described.

Soil Emanation Measurements

From the beginning of this work it was believed that the collection device and system should be designed to maintain conditions at the sampling site which simulated as nearly as possible those existing in the surrounding area during sampling. This requirement was extended to include preservation of antecedent environmental conditions of the sampling site as well. Because of this general requirement, a device to collect the gas being transferred into the atmosphere from the soil was designed. Also, a collection system was developed to separate the captured gas from environmental air for transportation to the laboratory where samples were processed.

The collecting device was designed to fulfill several requirements. First, minimal disturbance to the soil during the placement and operation of the collector was found to be essential. Also, the collector was to be constructed so that it could be sealed to the soil surface, thus prohibiting both loss of gases from the collector and the admission of external air where radon 222 content varies. Since it was attempted to perform measurements in a natural environment, flow within the collector was to resemble air movements in the free atmosphere. While providing such natural flow, the configuration to the collector had to be such that emanation could be reported in units of flux density (that is, quantity of gas emanating per unit area per unit time); thus, the area of soil being sampled had to be well defined by the collector. Finally, the device had to be durable, inexpensive, and easily fabricated.

The collector developed is shown in figure 2, in which the open bottom of the collector is visible by reflection. This opening covered an area of soil approximately 30.5 cm by 7.6 cm. Early models of the collector had sharpened edges about 2.5-cm deep which were pushed into the soil to seal the periphery of the collector to the soil. As undisturbed surface soils in central Illinois are mostly devoid of rocks and gravel, the seal was satifactory. However, the edges cut slots into the earth, permitting soil gases having relatively high radon contents to leak into the collector. Such a disturbance resulted in a large variation in measurements made only a few centimeters apart. Removing the knife edges from the collectors and sealing the collectors to the soil with caulking compound lowered the variation to a more satisfactory level and still afforded an acceptable seal. (With the knife edge seal, the standard deviation for eight samples taken close together and within a 1-hour interval was as high as 100 percent of the mean, while with the caulking material seal a 30-percent value was attainable on the same sampling sites.) The collector was designed as a straight tunnel in order to permit a reasonably constant flow across the entire surface being sampled.

After selection of an approximately flat surface, the collector was placed with the open face adjacent to the soil with minimum disturbance to the soil. As part of the consistent effort to maintain preexisting conditions at the site, the collecting device was carefully placed over the sampling area while the collecting system was in operation, thus ensuring that emanating radon could not accumulate within the system before the beginning of sampling. During the interval of sample collection, the device was not disturbed. At all times, operating procedures were such that flow across the soil surface simulated that existing in the unobstructed surrounding environment.

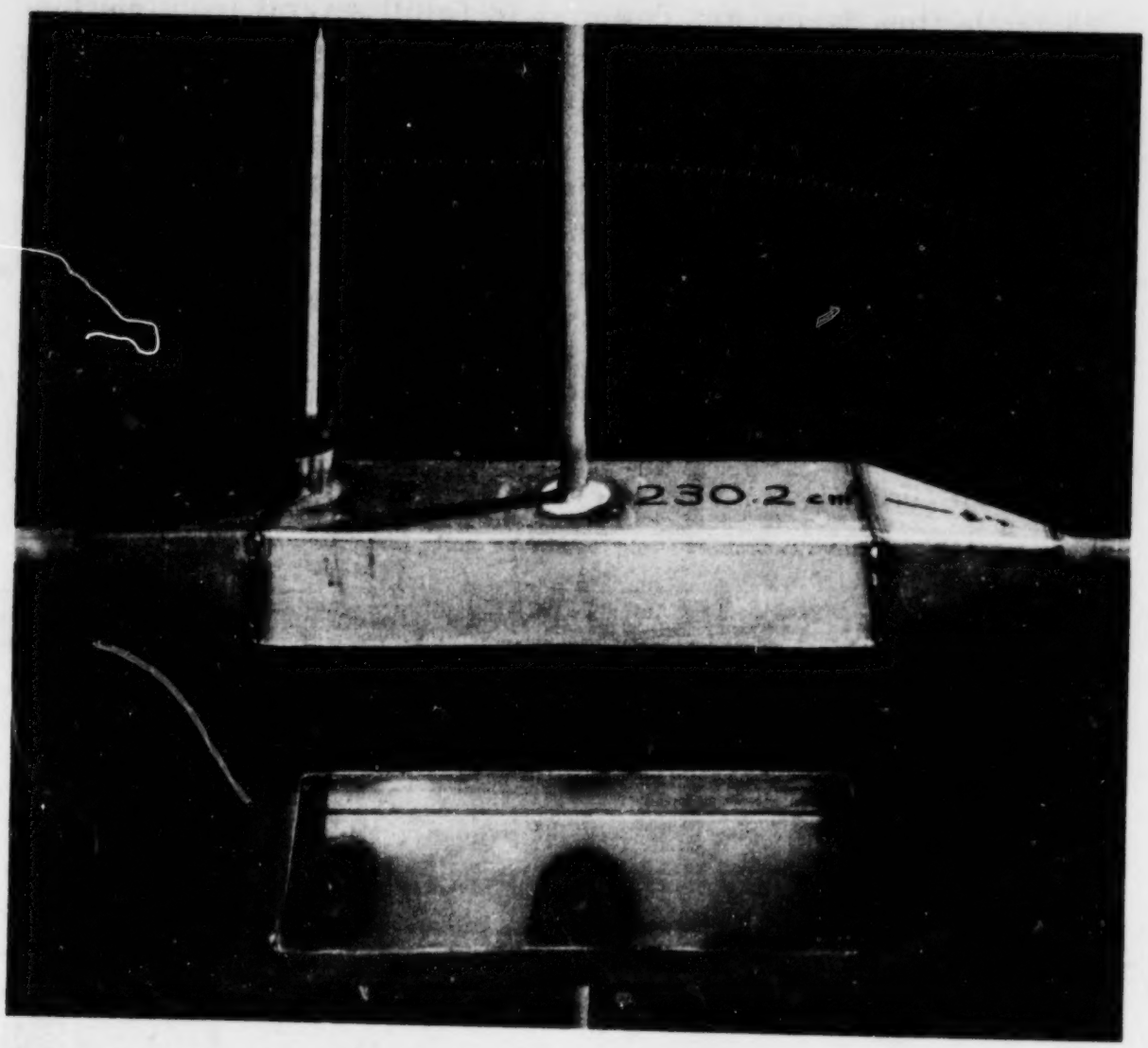

Figure 2. Collector for sampling radon 222 emanation at the earth-atmosphere interface. Opening in the collector is shown by reflection in a mirror.

The collector was fabricated from 16-gage sheet stainless steel which was formed and welded. This material provided sufficient stiffness for dimensional stability and, at the same time, satisfied the cost, durability, and ease of fabrication requirements.

The collection system to be used with the collector described above was designed to fulfill the criteria for sampling in the following respects. Sites where samples were to be taken were to be undisturbed for several days before sampling prohibiting the establishment of fixed installations which would interfere with rainfall, solar heating, and the turbulent structure of winds over the sites. The collection system also had to duplicate, as nearly as possible, the meteorological conditions over the area being sampled, and trouble-free operation of the system was a requisite for the successful accomplishment of uninterrupted sampling. These requirements were met by the use of a completely portable system by means of which flow within the collector could be adjusted to best simulate that in the free atmosphere.

The portable system designed included an alternating current motor generator; a plywood box containing a pump, tubing, valves, Dewar flasks, radon traps, and associated chemicals; a box for transporting shredded Dry Ice; and a box containing miscellaneous small supplies. The system is represented schematically in figure 3 and pictorially in figure 4. As mentioned above, the system was set in operation before placement of the collector. During the period between placing the collector and beginning the sample collection (2 or 3 minutes), the pump was run and the flow rate adjusted to give proper flow within the collector. Radon swept from the collector in this interval was removed from the closed-loop system by a flushing trap, which is a standard radon trap. At the initiation of sampling, the three-way valve was rotated 90° to start the collection of emanating radon in the sample trap. At the end of the sampling period (usually 300 seconds), a reverse turn of the three-way valve terminated the sample. At all times, then, flow within the collector was continuous.

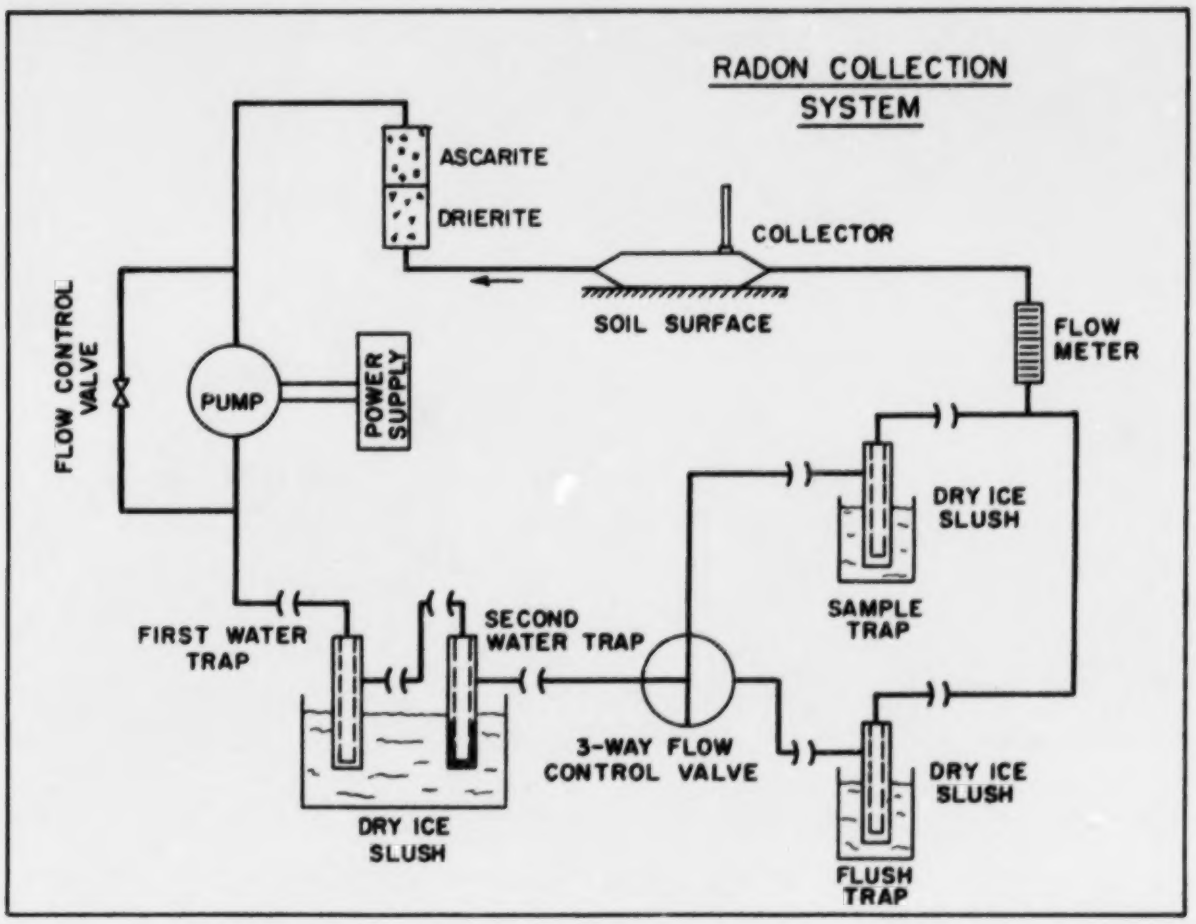

Figure 3. Schematic representation of the system for collecting radon 222 emanating at the earth-atmosphere interface.

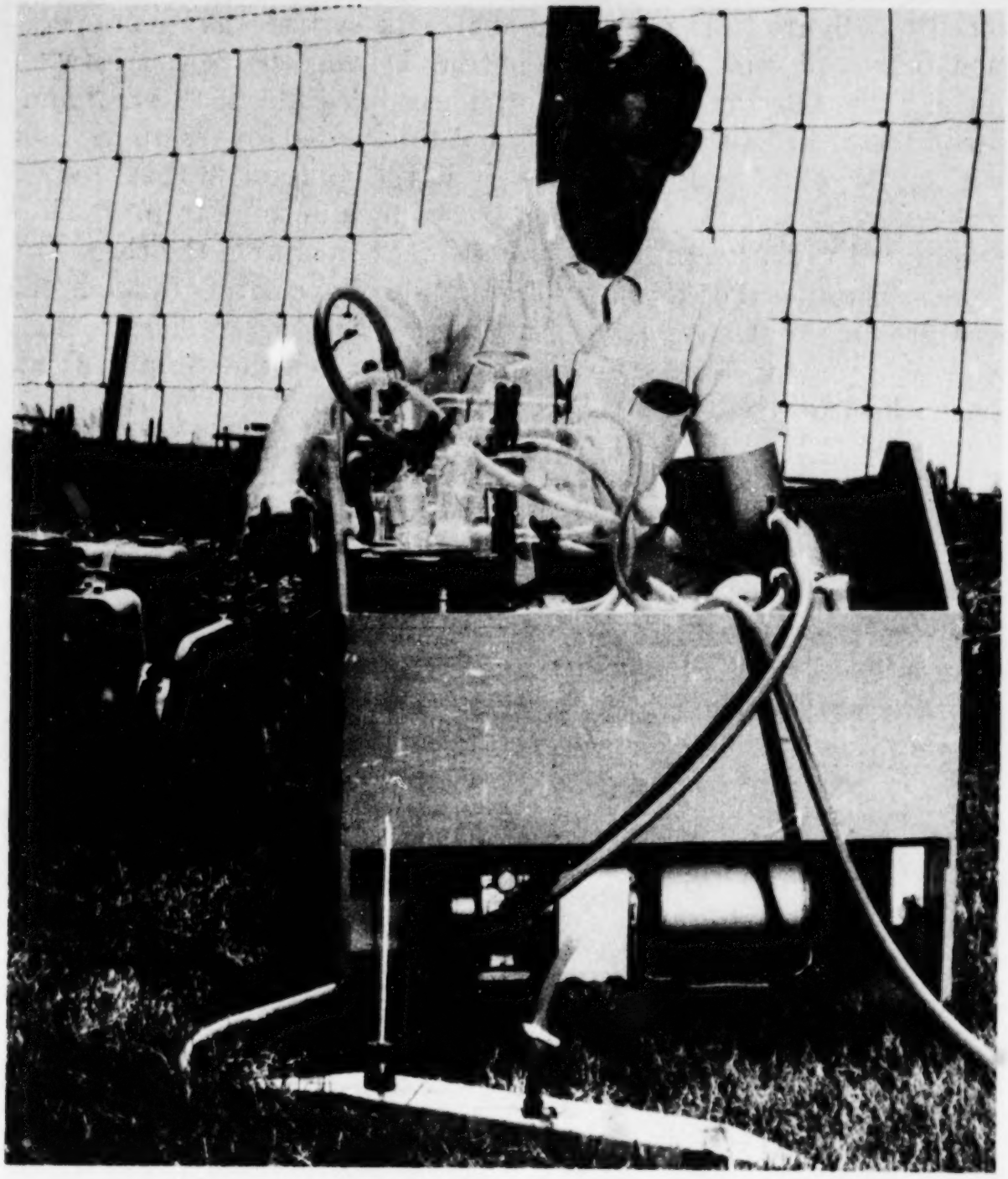

Figure 4. Portable system for collecting emanating radon 222 for use in the field.

MEASURED VALUES AND DISCUSSION

"Emanating Radium 226" in Soil Particles

In the presentation of a summary of this research, it seems logical to present first a summary of measurements made of the quantities of "emanating radium 226" in a variety of soils (table 2), as this is the source of the material being studied, radon 222, in the environment. In table 2, soils are classified, roughly, as those containing clays or fine materials, those which are predominantly sandy in texture, and those obtained at locations where uranium 238 and/or radium 226 are present in commercially important concentrations regardless of soil texture. The first three soil types listed, Flanagan, Catlin, and Drummer, cover approximately 85 percent of Champaign County, Ill., and are not statistically significantly different from each other (at the 90-percent-confidence level) with regard to "emanating radium 226" content.

It was observed that the soils with a sandy texture have a smaller "emanating radium 226" content than the soils of the clay type, except for the Zircon sand piled on the beach at Galveston, Tex. This latter material was used as a source of radioactivity during a cross-calibration experiment conducted in conjunction with the First International Symposium on the Natural Radiation Environment at Rice University, Houston, Tex., in April 1963. It is believed that the other sandy soils exhibited lower "emanating radium 226" concentrations than the clay type soils because sandy soil particles, being of greater size, have a smaller surface to volume ratio. Thus, a radon atom produced in the interior of a sandy soil particle has less chance of escaping into soil pore spaces, the measured "emanating radium 226" being therefore reduced.

"Emanating radium 226" contents of some of the soils from the mining areas, as expected, are higher than the others. Some of the samples obtained from Iceland were among the lowest activities of all those sampled. A sample of beach sand from Galveston, Tex., was lower. Throughout the various geological regions from which samples have been analyzed, observed concentrations of "emanating radium 226" varies about a hundredfold.

However, the ratio of maximum to minimum values for the clay type soils from the United States is only 2.3 : 1, and the ratio of values for the sandy soils, omitting the Zircon sand, is only 1.7 : 1.

Important studies of natural radioactivity of soils are presented in papers by Stout *et al.* (*6*), and Delwich *et al.* (*7*).

Table 2. Radioactivity of soils

[In 10^{-12} grams of "emanating radium 226" per gram of dry soil]

Location of soil type	Mixed clay soil	Sandy soil	Mining operation
Agronomy Plots, Urbana, Ill.			
Flanagan type (mean of 20 samples)	0. 679		
Catlin type (mean of 15 samples)	0. 633		
Drummer type:			
(mean of 15 samples)	0. 592		
(mean of 50 samples)	0. 64		
Argonne National Laboratory, Argonne, Ill. (mean of 3 samples)	0. 690		
Eddyville, Ill	0. 510		
Metropolis, Ill	0. 516		
Scottsville, Ky	0. 576		
Sylacauga, Ala	0. 794		
Albany, Ga. (red clay type)	0. 390		
Leesburg, Fla	------	0. 232	
Galveston, Tex. (beach sand)	------	0. 147	
Texas City, Tex. (Zircon sand piled on beach in 1963)	------	0. 623	
Rice University "Commons," Houston, Tex.	0. 460		
Atwood, Kans	0. 484		
"Windy Acres" near Liberal, Kans. (mean of 2 samples)	0. 484		
San Luis Valley, Colo	0. 342		
Central City, Colo. (mining area):			
Wood Mine (ore and tailings, respectively)	------	------	14. 3 and 0. 45
Xenotine from Spring Gulch	------	------	5. 26
Yttrium outcropping	------	------	2. 87
Blow sand over Todilito limestone, Grants, N.Mex	------	------	10. 4
Socorro, N.Mex., NMIMT:			
Weir Hall	------	0. 244	
Workman Mine (floor)	0. 414		
Iceland (6 locations)	0. 172		

Emanation of Radon 222 From Soil Beds

Table 3 presents a summary of a number of measurements of radon 222 emanation from soils. Again, Flanagan, Catlin, and Drummer soils were studied most carefully because they represent the most important soil source in the local area. At the New Mexico Institute of Mining and Technology, Socorro, N. Mex., measurements were made of soil emanation at the same site used by Wilkening and Hand (*8*). The results of both sets of observations are shown in table 3.

Table 4 lists individual measurements of the emanation of radon 222 at two locations where rock with high concentrations of uranium 238 outcrops the surface. The largest value observed near Grants, N. Mex., is approximately 1,000 times as large as that observed at many other locations. The large variation in values obtained was the result of sampling at varying distances from the outcrop over soil or blow sand of different depths.

Table 3. Rates of emanation of radon 222 from soils

[In 10^{-18} curies cm^{-2} sec^{-1}]

Location of soil type	Number of samples	Mean ± standard deviation
Champaign County, Ill.:		
Flanagan	72	111± (39)
Catlin	72	157± (72)
Drummer	72	151± (96)
Flanagan, Catlin, and Drummer	216	140± (73)
Brenton (5/11/64)	8	91± (25)
Sidell (5/11/64)	8	174± (42)
Thorp (5/15/64)	8	75± (18)
Proctor (5/15/64)	8	49± (11)
Cullo (5/15/64)	8	53± (16)
Argonne National Laboratory, Argonne, Ill. (1963)	8	57± (5)
Argonne National Laboratory, Argonne, Ill. (1965)	6	92± (30)
NMIMT, Socorro, N. Mex.:		
Weir Hall (Wilkening and Hand)	----------	90
Wier Hall (Pearson and Jones)	6	102± (30)

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Table 4. Rates of emanation of radon 222 from uranium 238 bearing rock outcrops over varying depths of soil or blow sand

[In 10^{-10} curies cm^{-2} sec^{-1}]

Sample number	Location	
	Central City, Colo.	Grants, N. Mex.
1	1, 358	1, 598
2	3, 154	21, 277
3	2, 280	21, 349
4	2, 444	127, 136
5	991	24, 932
6	563	43, 205
7	2, 870	48, 649
8	----------	13, 997
Mean and standard deviation	1, 951 ± (987)	37, 768 ± (26, 862)

Environmental Influences on Soil Emanation

The primary purpose of this study was to determine the effects of meteorological and soil conditions, time, soil type, and location upon the release of radon 222 from soils in the local area, Champaign County, Ill. These effects have been reported by Pearson *et al.* (*9*), Pearson and Jones (*10*), and are summarized herein.

WIND SPEED

While it was reasoned that conditions within the collection device should simulate natural air movements and gustiness, there existed a need to determine the relationship between wind speed across the surface of the soil and the emanation measured. First, a determination was made of the relation between flow rate within the collection system and wind speed within the collecting device measured at 1.25 cm above the soil using an anemometer operating on the direct-heat themopile principle. A variety of flow rates were used while the concomitant emanation was collected and determined. The results are shown in figure 5 in which the linear relationship, at the wind speeds studied, is apparent. Knowledge that there is a dependency of emanation upon wind speed across the soil surface led to the conclusion that wind speeds existing just above the soil in the surrounding area should be measured and duplicated as closely as possible within the collector during sampling.

Wind speeds in the free atmosphere were measured at 1.25 cm above ground level using the hot-wire anemometer previously mentioned. Average speeds of 0.03 m/sec and 0.08 m/sec were measured for an

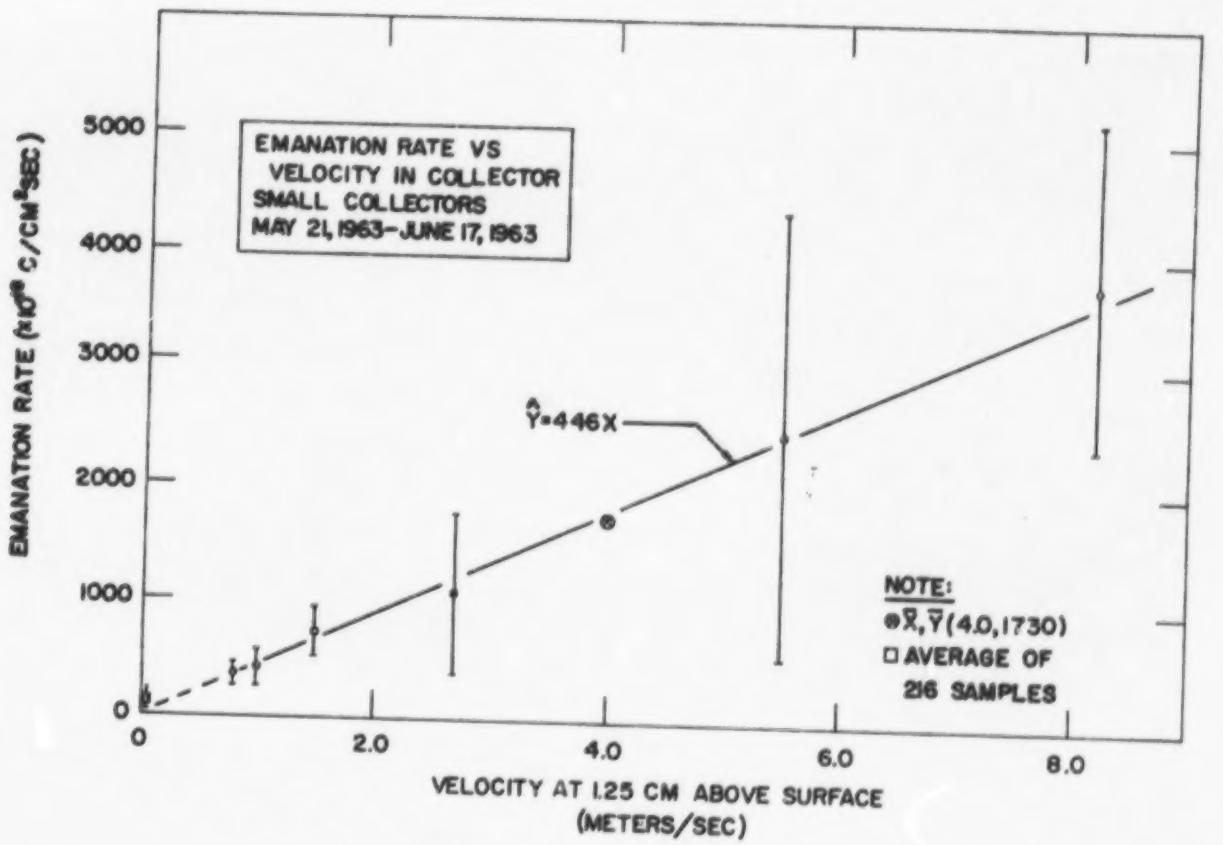

Figure 5. Emanation rate of radon 222 as a function of wind speed within collector.

average and a windy day, respectively. The results of the testing showed that a flow rate of 4 liters/min in the system would produce, within the collecting device, a measured velocity of 0.045 m/sec at 1.25 cm above the ground, a speed approximating those occurring naturally almost all of the time. It should be noted in figure 5 that the average for 216 samples taken at the flow rate of 0.045 m/sec does not fall on the dotted line. Also, the emanation measurements reported by Evans *et al.* (*11*) were taken with a wind speed of zero and are not greatly different from the mean of 216 samples. It is concluded that for the very low wind speeds, ordinarily observed between the grass blades at 1.25 cm above the ground, there is no demonstrable variation of emanation with variation in wind speed. The left end of the curve in figure 5 should become approximately horizontal at the very low wind speeds which normally prevail.

Gustiness patterns were observed on the wind speed meter with the anemometer probe at 1.25-cm elevation in the free atmosphere, and with the probe at the same elevation inside the collector with a flow rate of 4 liters/min. The range and frequency of pulsations appeared similar in both cases.

It was decided, as a result of these tests, to adopt a collection flow rate of 4 liters/min provided this flow could be shown to be satisfactory

by a study of its effects on emanation rate over a period of several hours. This was ascertained by placing the collector over a fixed location and sampling continuously for an 8-hour period. Samples were collected in a charcoal sampling trap for 55 minutes. Then the flow was diverted into one of the flushing traps for 5 minutes while the sampling trap was removed from the system and replaced with a freshly purged one. This cycle was repeated each hour. Eight-hour tests of this design were carried out on two different days. Data obtained are presented graphically in figure 6. While natural conditions prevailing during the two testing periods established substantially different emanation rates, the uniformity of measurements within each test indicated that the removal of radon at the surface, while using the adopted flow rate, neither caused a depletion of radon from the soil nor permitted it to build up in concentration just below the surface. The former result would be evidenced by a decreasing emanation rate with time and the latter by an increasing rate. Neither situation was observed.

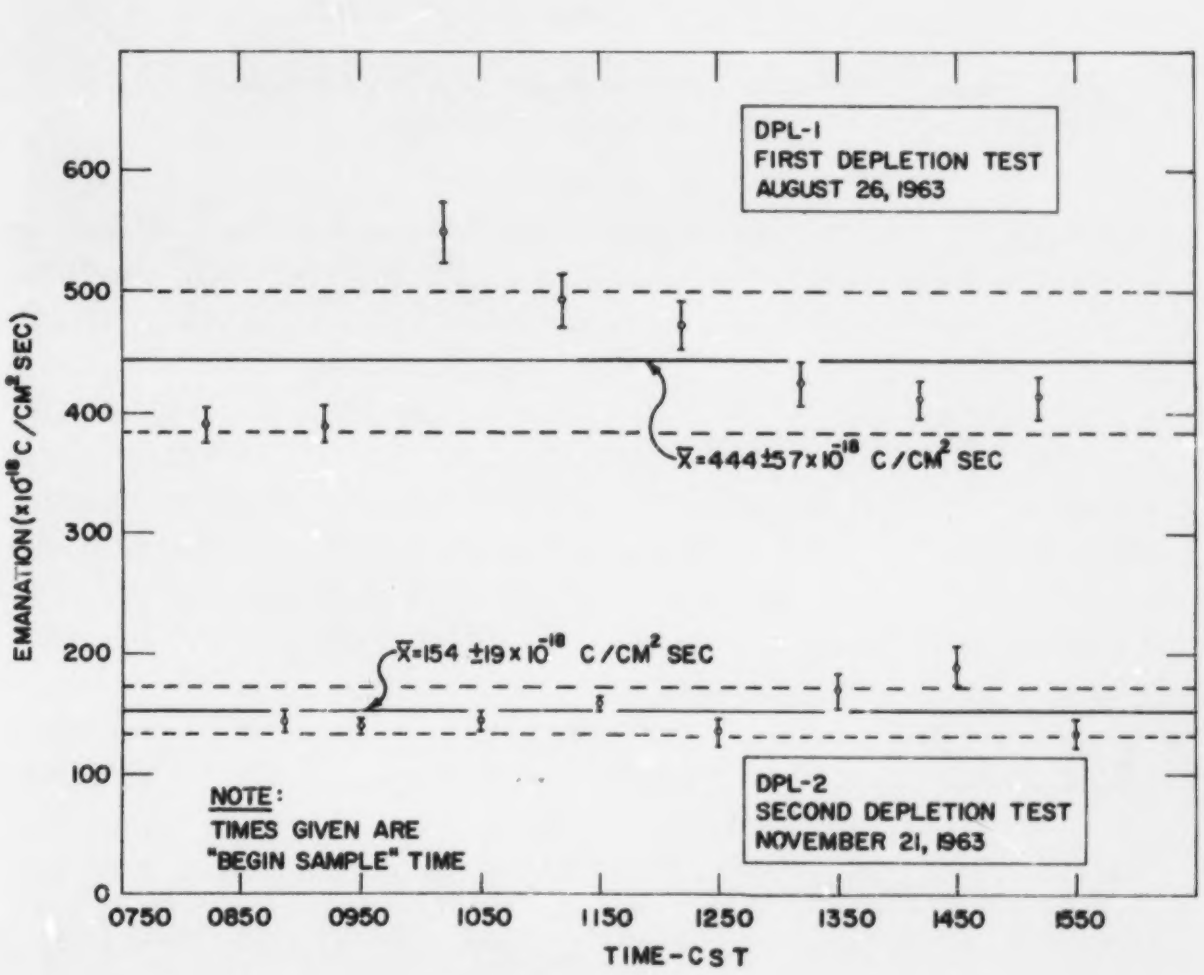

Figure 6. Results of experiments to determine the suitability of adopted flow rate (4 liters/min) within collector. Upper data from tests of August 26, 1963; lower date from that of November 21, 1963. Curves show the means ±1 standard deviation for each of the two tests.

SOIL MOISTURE

When soils were moistened with approximately 25 mm of water and observed during the following 8 hours of drying, no significant variation of emanation with soil moisture was observed. However, for greater lengths of time, days to weeks, a significant variation of emanation with soil moisture was observed. When the soil was wet (air void fraction [1] about 0.17), the emanation measured was approximately one-third of that observed when average moisture conditions (air void fraction about 0.40) were prevalent.

DIURNAL EFFECTS—ATMOSPHERIC TURBULENCE

Emanation was observed to vary markedly in one 24-hour study. The variation of emanation was attributed primarily to atmospheric turbulence. Thus, near sunrise and in midafternoon, when the atmosphere was most turbulent near the soil surface, radon effluxation was highest and most erratic. During the stable nighttime, the flux was relatively constant, increasing slightly throughout the night. This increase was attributed primarily to restoration of the radon concentration in the top layers of soil which were depleted during the previous daytime period of high emanation.

SEASONAL EFFECTS

There was a marked decrease of emanation when soil was cold or frozen. However, a very high and erratic emanation was observed during a January thaw when approximately 7 cm of soil nearest the surface were not frozen.

LOCATION AND SOIL TYPE EFFECTS

A study of nine sites within a 13-km radius from the University of Illinois, Urbana, and over the three most common local soil types revealed, at the 90-percent-confidence level, no differences in emanation owing to location or soil type (*10*). As soils in the prevailing upwind direction (for hundreds of km) have the same geologic history, it is believed that the local site is suitable for studies of vertical diffusivity using radon 222 as a tracer material. However, at a few sites, in the uranium-radium producing areas, emanation approximately 1,000 times that of ordinary soils was observed.

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[1] Air void fraction is used to indicate that fraction of a soil volume occupied by soil gas.

BAROMETRIC PRESSURE, ATMOSPHERIC TEMPERATURE, AND RELATIVE HUMIDITY

Barometric pressure, atmospheric temperature, and relative humidity also may have produced some response, but the data are too limited and the effects too small to permit any definitive statements.

Leaf Emanation Measurements

The water-air phase distribution ratio of 0.25 at 20° C. for gaseous radon (*12*), and the 100 to 1,000 times greater concentration of radon in soil gas as compared to the atmosphere above the soil (*11*), suggested that soil water would be relatively rich in dissolved radon. From these two conditions, the hypothesis was forwarded that plant roots acquiring the radon 222 rich soil water subsequently transport both water and dissolved radon to above-ground plant parts where both are released to the atmosphere. An investigation of such phenomena was accomplished in cooperation with Dr. Richard R. Dedolph, Division of Biology and Medicine at Argonne National Laboratory, Argonne, Ill.

The soil emanation measurement system was modified so that leaf radon 222 emissions could be determined under nearly natural conditions. These sampling modifications provided, within the collecting device, nearly normal light intensities, as well as movement of air, with nearly normal concentrations of water vapor and carbon dioxide. Six measurements of soil emanation and six measurements of release of radon 222 from leaves of field corn (*Zea mays L.*) were made on August 5, 1966, at Argonne National Laboratory, when the corn was beginning to develop tassels. These measurements were made between 1100 and 1600 CST. At this time, release from leaves was measured at $(247 \pm 19) \times 10^{-18}$ curies cm^{-2} sec^{-1}. While the leaf to soil release ratio was over 2.6:1 for this experiment, it should be noted that the soil emanation was somewhat low, probably because of the high soil moisture content. However, the leaf contribution was greater than the average soil emanation for Champaign County, Ill., 140×10^{-18} curies cm^{-2} sec^{-1}. This coupled with the fact that the corn leaf area to soil area can reach the ratio of 3:1 emphasizes the importance of considering the radon 222 contribution of plants during daylight hours.

Atmospheric Concentrations of Radon 222

While previously reported studies provide insight into the mechanism of release of radon 222 to the atmosphere, primary interest centers on the concentrations of that gas which accumulates in the

environmental boundary layer of the atmosphere. Studies of concentrations of radon 222 in the atmosphere have been made in cooperation with Mr. H. Moses at the Argonne National Laboratory, Argonne, Ill., from the surface of the earth to 16-m elevation for eight periods of approximately 24 hours each using the sampling method described by Moses *et al.* (*13*). The data include a total of 650 observations of radon concentrations in the atmosphere. Concurrently, meteorological measurements were made at or near the sampling site.

While the data provide an insight regarding the variables, it is recognized that the information is limited by (1) the small number of samples, (2) the use of only one geographical location, (3) the collection of samples during a limited time—the late spring and summer, and (4) the limited height of the atmospheric layer above the earth under consideration.

The surface of the earth near the sampling site (within 30 m) is fairly level; the slopes are gradual; elevation changes are limited to ±2.1 m, except for a steep descent toward a ravine to the southwest. The 300-m circle is almost without trees; a small clump of 6-m-high saplings stands in a shallow hollow about 38 m to the north, and a line of similar trees runs down the slope to the southeast. In the southwesterly direction, the nearest trees, full-grown oaks, are 380 m to 430 m away. The ground cover of coarse grass and weeds was mowed regularly and was 4 cm to 16 cm in height within 30 m of the sampling site. At 90 m southwest of the site was a 46-m tower of open steelwork construction and at about 185 m west was the Meteorology Building, a gable-roofed structure, 4.3-m high by 9-m wide, as viewed from the site.

Elevations at which radon samples were taken, for almost all of the tests, include one between the surface and 1 cm above the surface, and 0.5, 1.0, 4.0, and 16.0 m above the ground. All measurements or samples reported represent mean values for 20-minute sampling periods centered at the reported time.

Although eight 24-hour periods of data represent a rather limited number of samples, the eight periods represent a variety of normally occurring meteorological regimes. At one extreme are cloudless conditions with strong incoming radiation during the day and pronounced outgoing radiation at night producing a marked inversion, while at the other is an overcast situation with greatly attenuated incoming radiation during the day with strong winds, good vertical mixing, and weak or no inversion conditions at night. It is expected that a majority of the conditions observed during spring and summer periods are between these extremes.

DISTRIBUTION OF ATMOSPHERIC RADON CONCENTRATIONS UNDER TWO DIFFERENT METEOROLOGICAL REGIMES

The cloudless situation is represented by the data for July 28–29, 1960, in figures 7 and 8, while the cloudy regime with strong winds is represented by the data for June 23–24, 1960, in figures 9 and 10. Figures 7 and 9 show radon concentrations on a height-time plane, while figures 8 and 10 show the associated meteorological conditions.

The two radon concentration diagrams, figures 7 and 9, have several common characteristics. Highest concentrations are observed next to the ground, the source. Maximum concentrations occur during the night and minimum concentrations during the day. Late morning radon concentration profiles show little variation with height, but in the early evening, concentrations increase more rapidly near the ground than above as shown by the slanting isopleths.

Figure 7 exhibits greater concentrations at all times and elevations than does figure 9. In figure 7 the maximum concentration at the ground was observed at 0230 CST while the maximum at 16 m occurred 3 hours later, shortly after sunrise. This illustrates the inverse fumigation condition (*13*). The weakest concentrations were observed at about 1300 CST. While the concentrations in figure 9 may be attributed in part to the strong winds with pronounced mechanical turbulence and good vertical mixing of materials in the lower atmosphere, they may also be due to a reduced rate of emanation of radon 222 from the soil owing to the presence of soil moisture. Rain fell for 3 days preceding this test. On the morning of June 20, 1960, 0.05 inch of rain fell. In the early afternoon of June 21, 1960, another 0.05 inch of rain was added, while on June 22, 1960, steady rain from 0900 to 1500 CST provided an additional 0.20 inch. It has been suggested, also, that rising atmospheric pressure will inhibit release of radon (*11*). Figure 10 shows this to be occurring during the June test.

Maximum concentration of radon 222 reached near the surface on the June 23–24, 1960, test was 0.732×10^{-12} curies liter^{-1}. Also, the daylight minimum of the June test occurring at 0700 CST was less than one-half of the minimum for the other test occurring near 1430 CST.

AVERAGE TIME-HEIGHT RELATIONSHIPS TO RADON CONCENTRATION

In the preceding section, data taken in two individual series are presented. In this section, averages of eight sets of samples, each taken over about 24 hours, are considered. Five of the 8 days when tests were run were similar to the one presented in figures 7 and 8, that is, high incoming radiation during the day, and high outgoing radiation at night, while three tests were similar to those shown in figures 9 and 10,

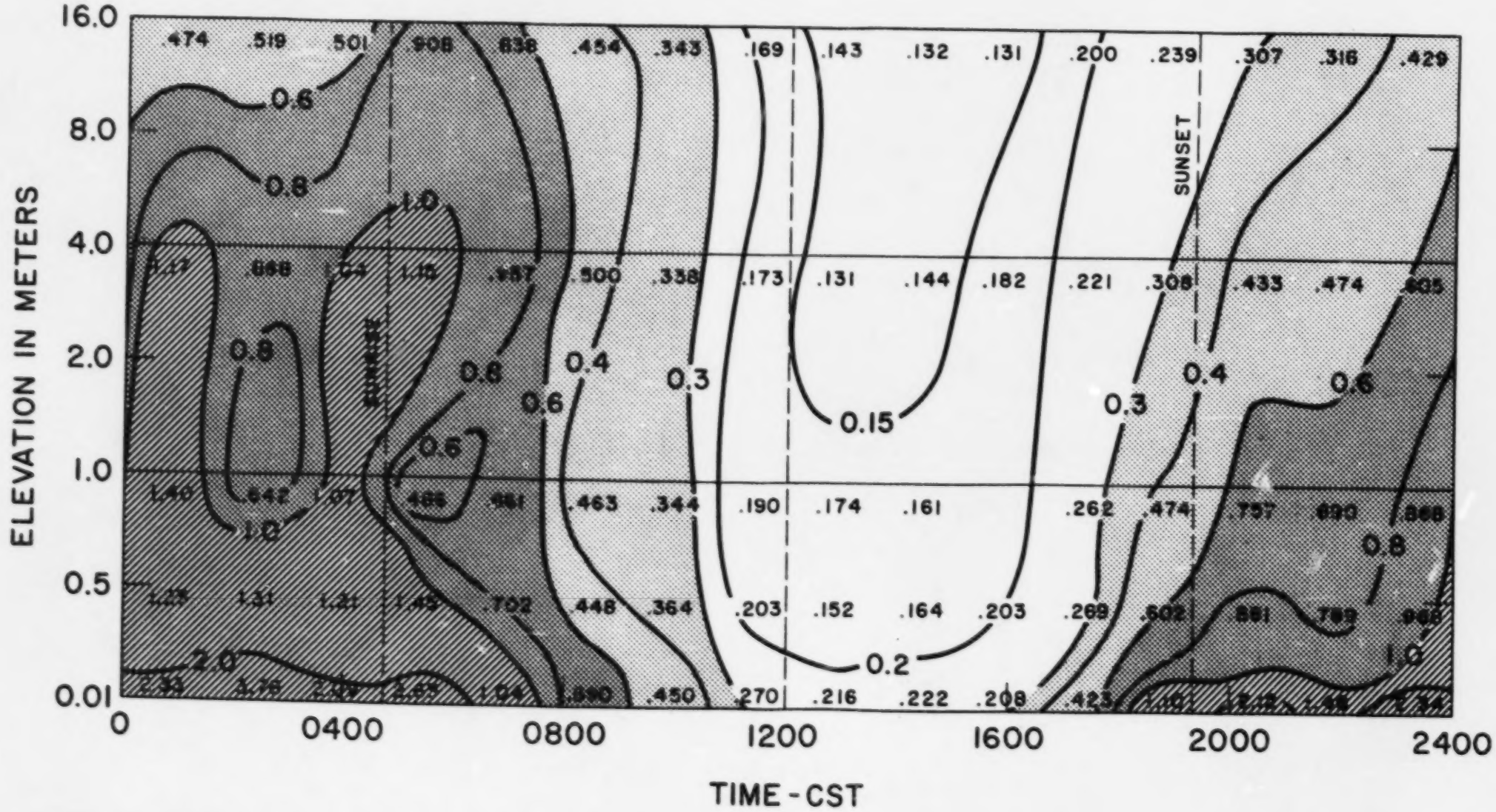

Figure 7. Radon 222 concentrations (in 10^{-12} curies liter^{-1}) variation with elevation and time for July 28–29, 1960, a period with scattered to overcast clouds during the day and clear skies, light winds and inversion during the night.

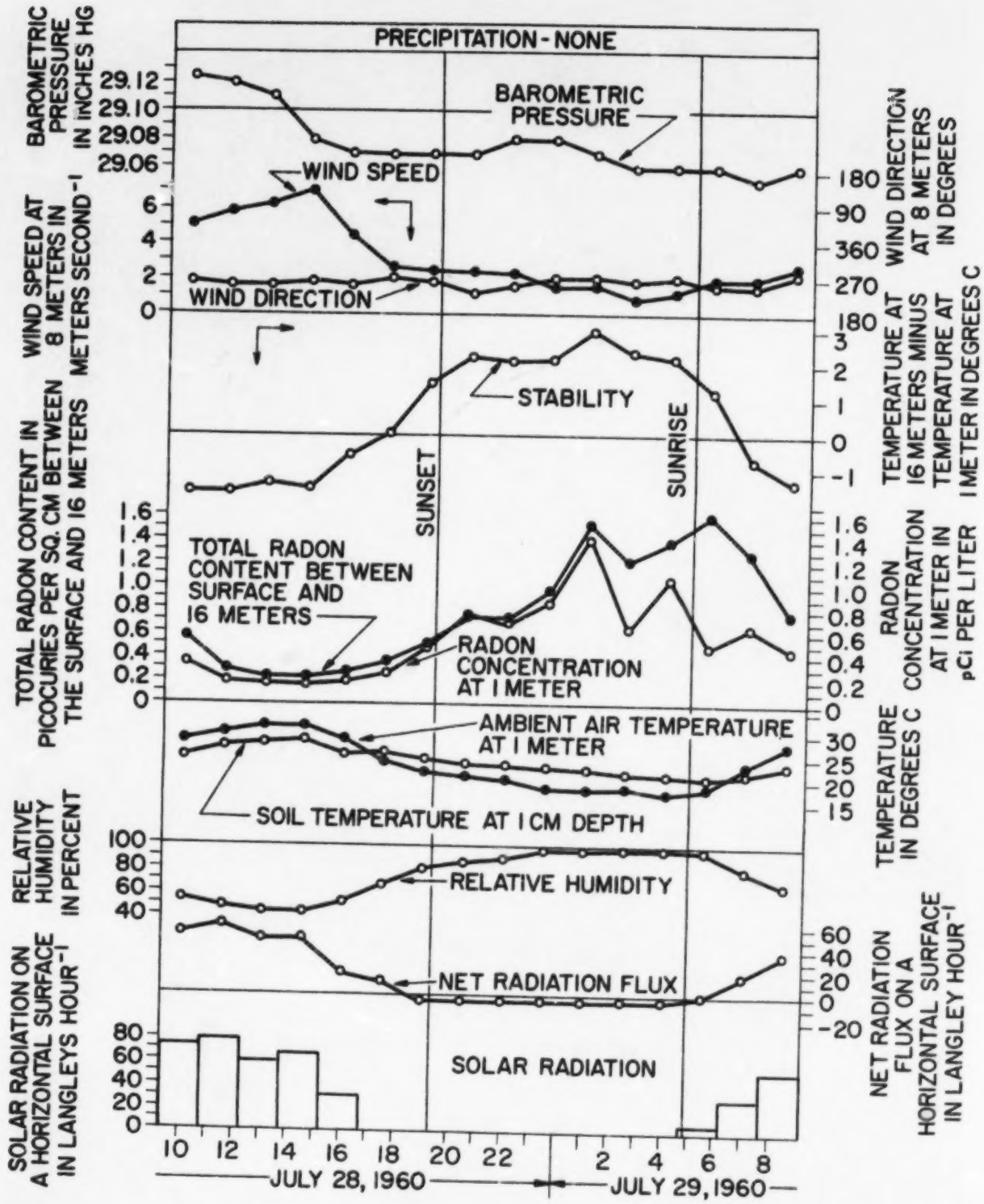

Figure 8. Radon 222 concentrations and related meteorological variables for July 28–29, 1960.

where high wind conditions and cloudiness resulted in distributions of relatively low radon concentrations. Such a body of data may have a bias toward the clear day regime and is not presented as typical of a climatic average of radon. However, the data give an indication of radon behavior in the atmosphere and are presented with recognized limitations.

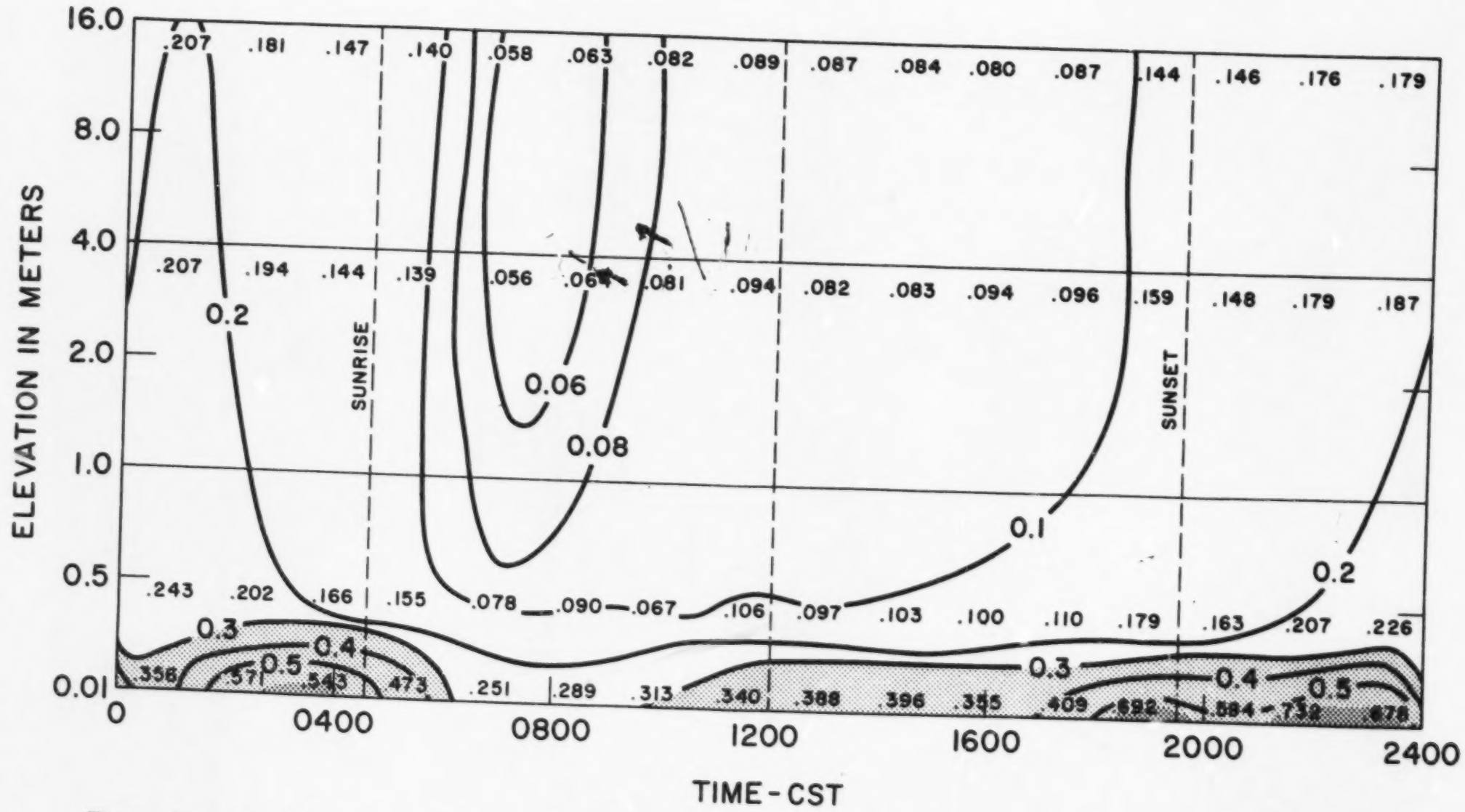

Figure 9. Radon 222 concentrations (in 10^{-12} curies liter^{-1}) variation with elevation and time for June 23–24, 1960, a period with overcast skies during daylight and relatively high wind speeds with a weak inversion at night.

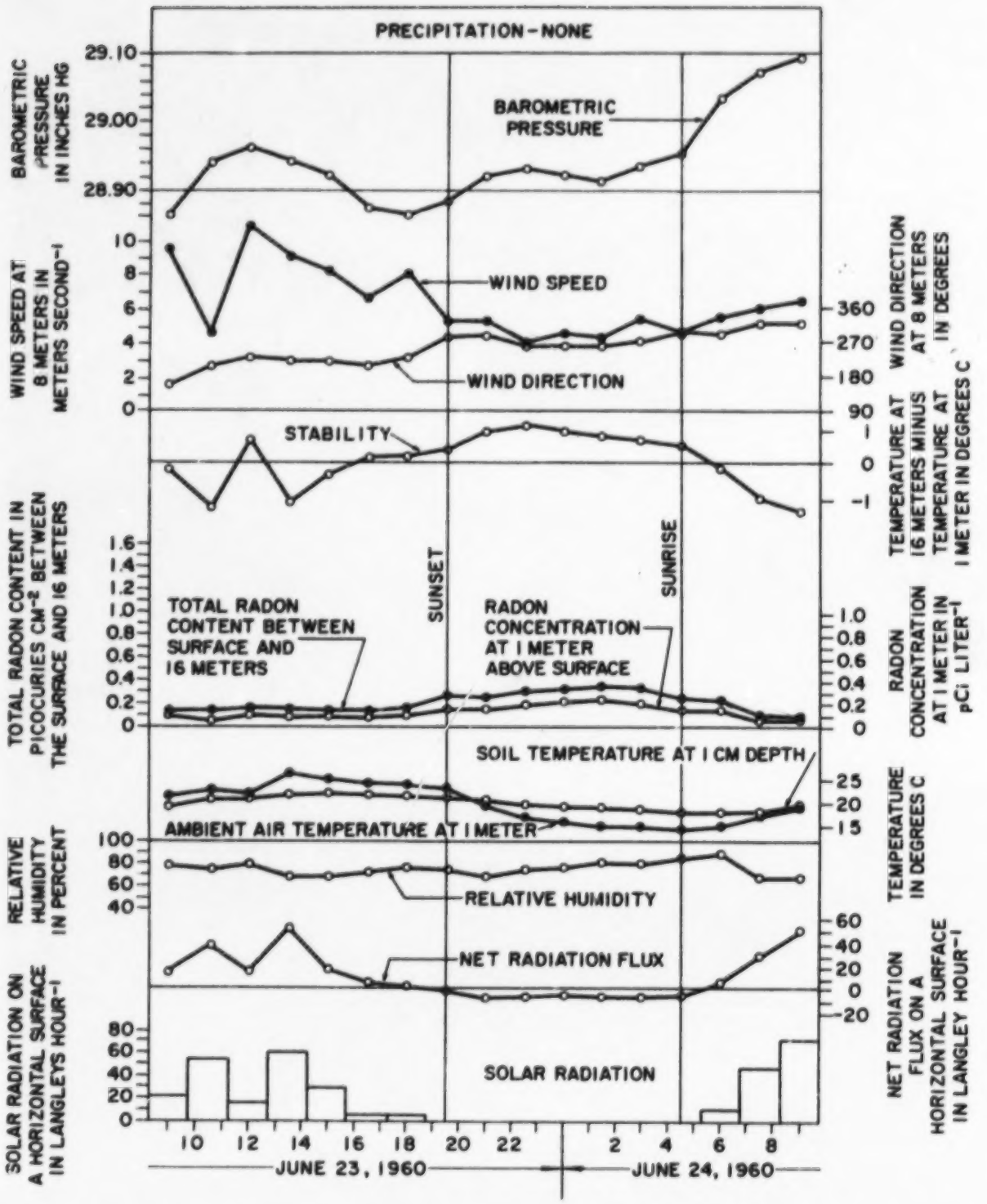
PRECIPITATION - NONE
BAROMETRIC PRESSURE IN INCHES HG
29.10
29.00
28.90
BAROMETRIC PRESSURE
WIND SPEED AT 8 METERS IN METERS SECOND⁻¹
WIND SPEED
WIND DIRECTION
WIND DIRECTION AT 8 METERS IN DEGREES
STABILITY
TEMPERATURE AT 16 METERS MINUS TEMPERATURE AT 1 METER IN DEGREES C
TOTAL RADON CONTENT IN PICOCURIES CM⁻² BETWEEN THE SURFACE AND 16 METERS
TOTAL RADON CONTENT BETWEEN SURFACE AND 16 METERS
SUNSET
SUNRISE
RADON CONCENTRATION AT 1 METER ABOVE SURFACE
RADON CONCENTRATION AT 1 METER IN pCi LITER⁻¹
SOIL TEMPERATURE AT 1 CM DEPTH
AMBIENT AIR TEMPERATURE AT 1 METER
TEMPERATURE IN DEGREES C
RELATIVE HUMIDITY IN PERCENT
RELATIVE HUMIDITY
NET RADIATION FLUX
NET RADIATION FLUX ON A HORIZONTAL SURFACE IN LANGLEY HOUR⁻¹
SOLAR RADIATION ON A HORIZONTAL SURFACE IN LANGLEYS HOUR⁻¹
SOLAR RADIATION
10 12 14 16 18 20 22 2 4 6 8
JUNE 23, 1960
JUNE 24, 1960

Figure 10. -Radon 222 concentrations and related meteorological variables for June 23–24, 1960.

If average concentrations, shown in figure 11, for the eight tests are presented in the same fashion as in figure 7, essential features are quite similar. These features include: (1) the high buildup in the early morning hours near the ground, the inverse fumigation just after sunrise, (2) the vertical lines representing vertical concentration gradients or very good mixing during the morning hours, (3) the minimum concentrations around noon or shortly thereafter, and (4) the evening lag in the increase of concentration in the upper levels after it has occurred near the ground resulting in sloping isopleths, as may be seen in the right side of figure 11.

Figure 12 represents the same data, but the average radon concentration is presented as the ordinate and time as the abscissa. The isopleths are lines of constant elevation. Figure 12 points out the time rate of change of radon concentration at various times of day. Also plotted on this figure are average values for each level based on all of the data available. It is clear from this figure that average radon concentrations at the breathing level, that is, about 5 feet, if measured during the daytime, would provide values that were appreciably lower than those for the total 24-hour period or for the night hours.

In figure 13 radon 222 concentration is presented as a function of height for selected hours. Observations lying between those shown in the figure were omitted to avoid confusing clutter. The series of times presented does not include the daytime and nighttime extremes, which are shown on figure 14. The dashed line with squares in figure 13 represents the average of all the data for each level, and therefore represents an average profile of radon concentration. Besides showing the vertical gradient, it shows quite markedly that the diurnal spread in values of radon concentration is large in the lower levels and converges to a substantially smaller range at 16 m. Each point shown in this figure represents an average of approximately eight observations. Thus, figures 11, 12, and 13 emphasize different features of the radon-time-height distributions, and taken together provide a more complete picture of the interrelationships involved.

The curves in figure 14 represent eight test averages for the times when the lowest and highest concentration profiles were observed—1430 CST—when radon concentration was at a minimum, and 0230 CST when concentrations are relatively high. Each point shown in figure 14 represents a single measurement at the time indicated. The dispersion of the data and the difference in dispersion of data points for the two times presented are interesting features of figure 14.

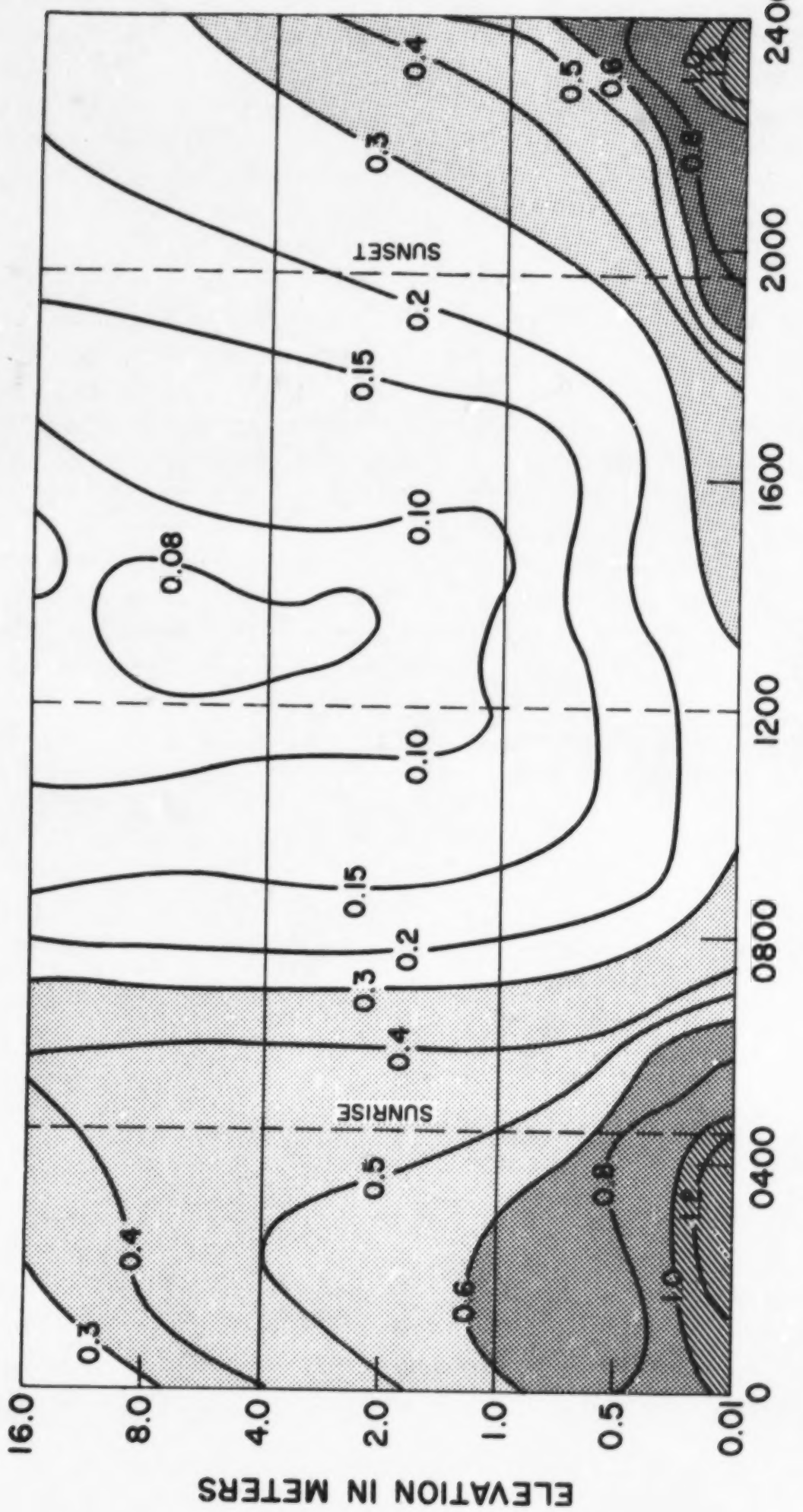

Figure 11. Mean radon 222 concentration (in 10^{-13} curies liter^{-1}) variation with elevation and time for eight 24-hour periods.

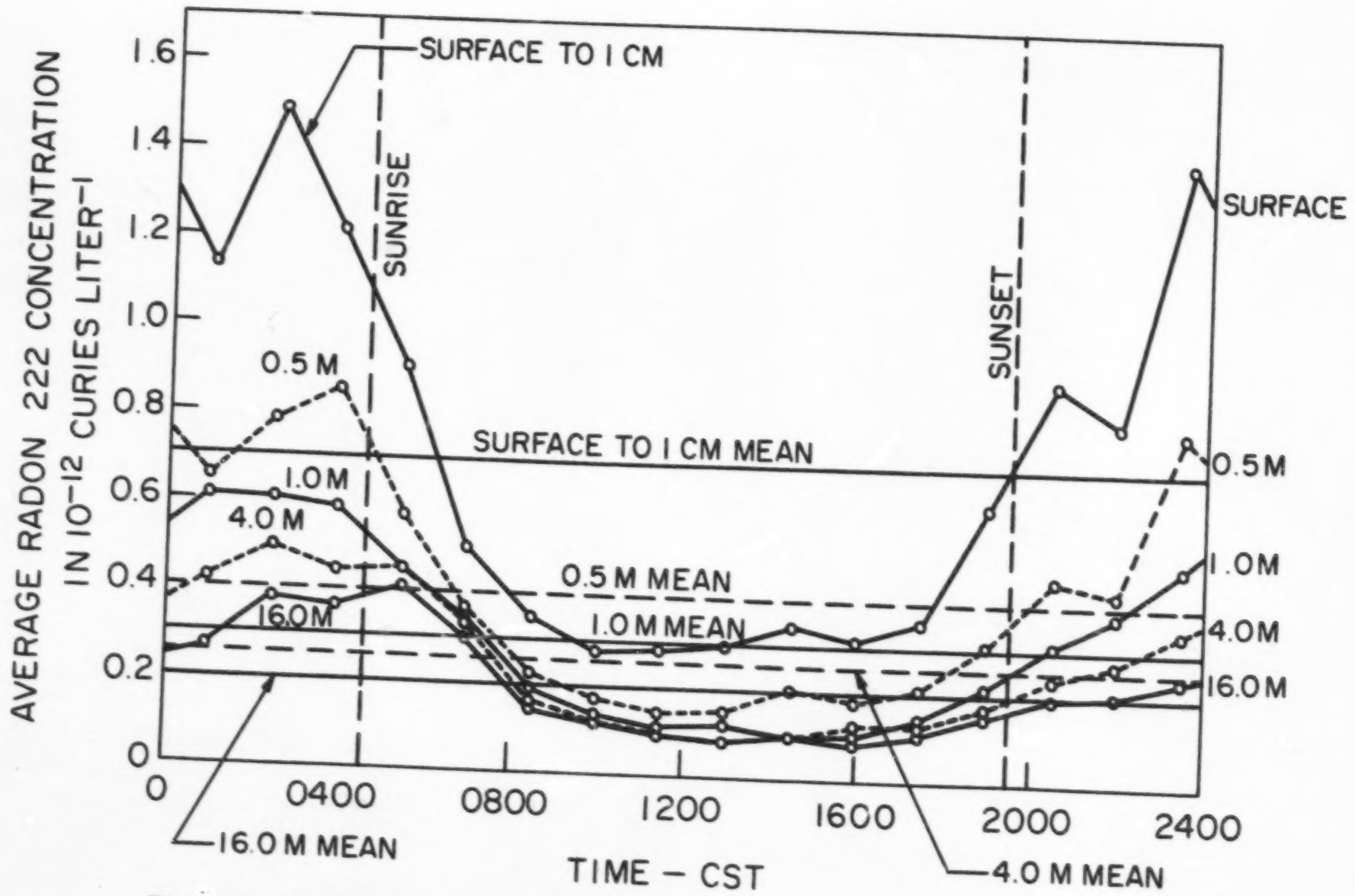

Figure 12. Variation of mean concentration of radon 222 with elevation and time for eight 24-hour periods and average 24-hour concentrations.

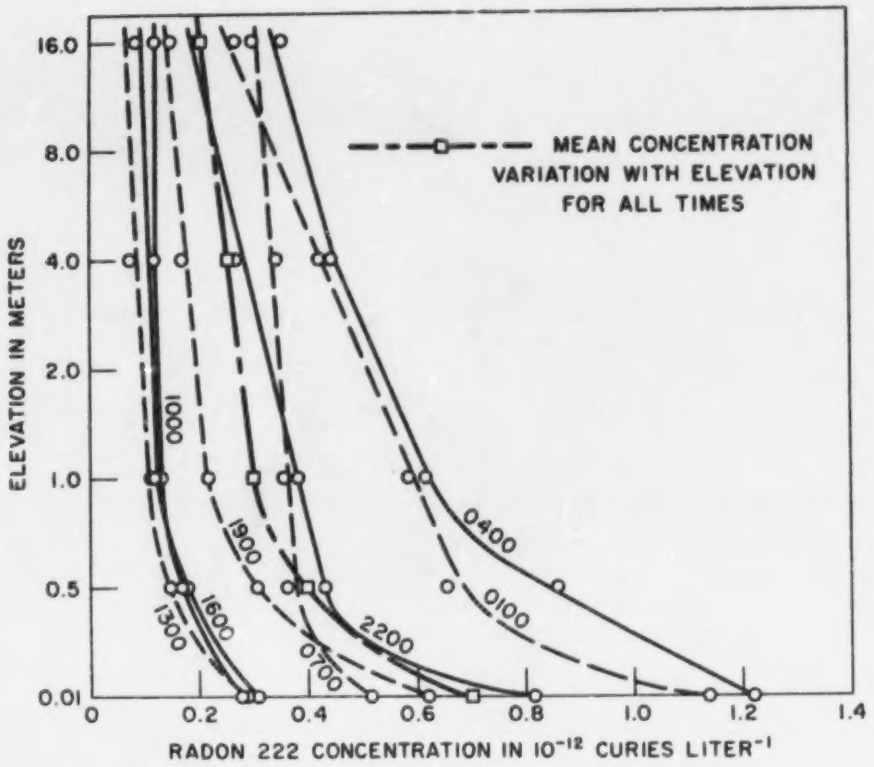

Figure 13. Average radon 222 concentration variation with time and elevation and mean concentration variation with elevation for all times (CST).

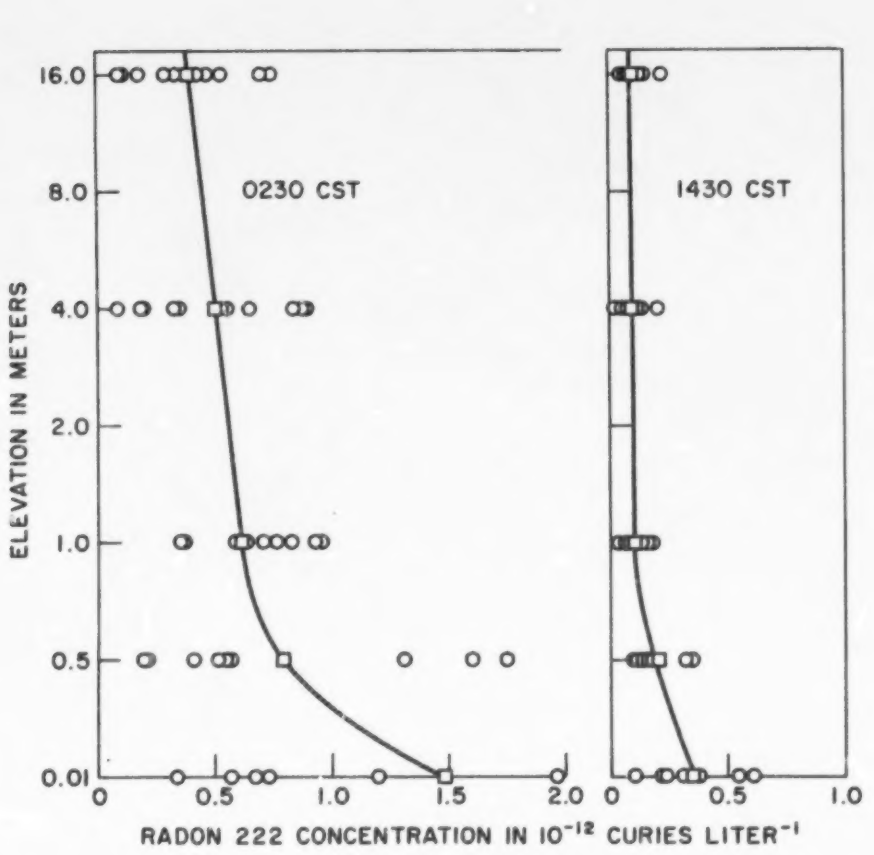

Figure 14. Individual radon concentrations measured at various elevations at 0230 and 1430 CST and mean curves for the times indicated.

FREQUENCY DISTRIBUTION OF RADON CONCENTRATIONS AT VARIOUS HEIGHTS

Figures 15 and 16, further illustrating how radon varies with height, are frequency distributions of observations for the daylight hours, 0500 to 1830 CST, and the nighttime hours, 1830 to 0500 CST. These figures show the large variability of radon concentrations near the surface as compared with those at 4 m or 16 m. Also, the value of radon concentrations at the mode decreases with height. Values of the standard deviation for each level for daytime and nighttime values are shown in table 5. As is evident from the data, the standard deviation tends to decrease with height, both during day and night, but the daytime values are considerably smaller. Table 5 also summarizes the measurements by giving values for the absolute maximum, average of the maxima for the eight series of observations, the average of all the data, average minima for the eight values, and the absolute minimum for each of the levels of measurement. Of course, during the day with solar heating, vertical motions are intense enough to provide mixing over a relatively thick layer. During the night, with the presence of inversions, this mixing is suppressed and allows the radon or any material emanating from the ground or having its source near the ground to become concentrated in the lower layers. The increase in the lower layers is shown quantitatively by these data.

Table 5. Radon concentrations (pCi/liter) at various elevations at Argonne National Laboratory in late spring and summer, 1958 and 1960

Type of data	Concentrations (at various elevations)				
	0.01 m	0.05 m	1.0 m	4.0 m	16.0 m
Maximum observed	3.930	2.429	1.400	1.170	0.908
Average maximum	1.732	0.957	0.764	0.570	0.449
Overall average	0.705	0.407	0.301	0.259	0.204
Average minimum	0.177	0.101	0.084	0.090	0.078
Minimum observed	0.103	0.067	0.032	0.018	0.019
Daytime mean and standard deviation	0.385±0.214	0.234±0.168	0.181±0.153	0.179±0.197	0.164±0.164
Daytime median	0.33	0.18	0.14	0.11	0.11
Nighttime mean and standard deviation	1.065±0.869	0.636±0.534	0.454±0.297	0.339±0.243	0.261±0.154
Nighttime median	0.72	0.46	0.38	0.27	0.22

Admittedly, the information provided in this summary is far less than necessary to provide a complete picture. Nevertheless, conditions representing high diffusion rates and the low diffusion rates discussed here bracket the majority of weather conditions which may occur. Thus, this analysis may be used as a guide concerning distributions of radon as well as those of other contaminants, such as pollens or auto exhausts, which may be injected into the lower atmosphere. The same atmospheric processes operate in all of these cases.

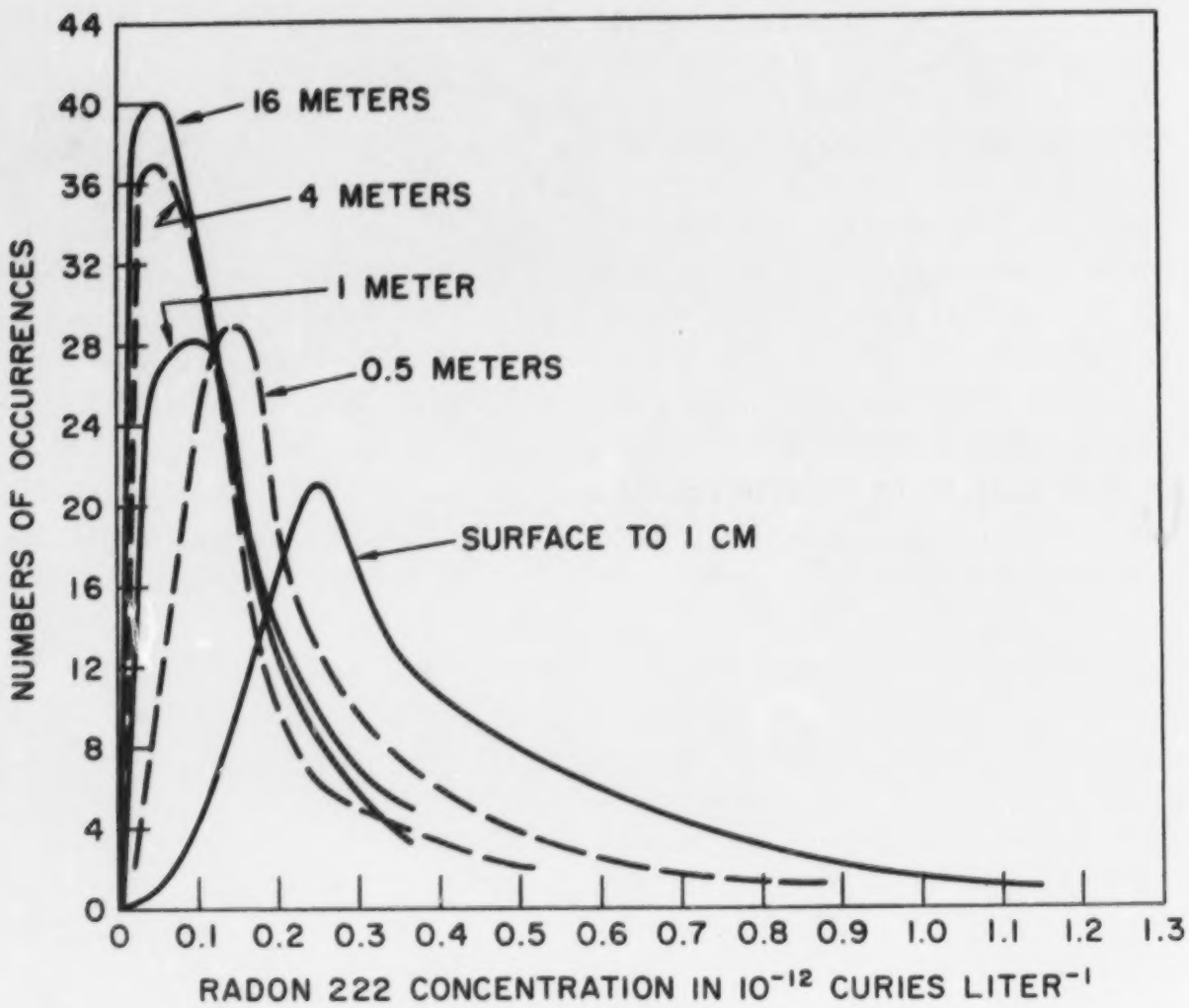

Figure 15. Frequency distributions of daytime radon 222 concentrations from 0500 to 1830 CST.

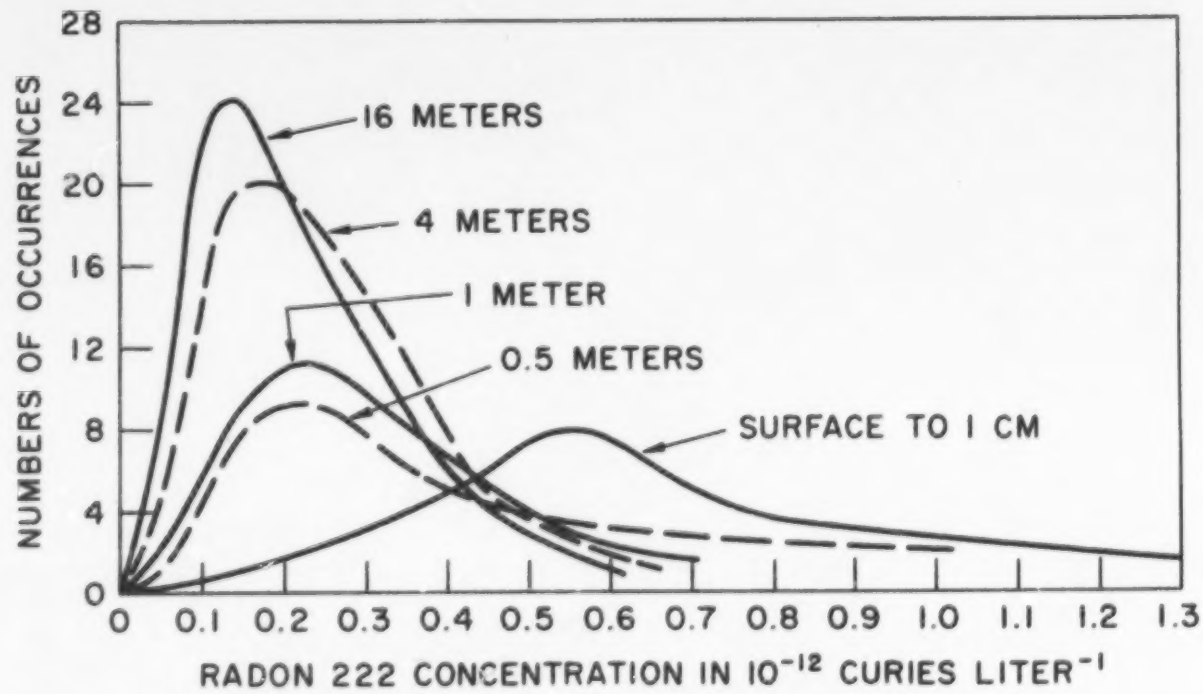

Figure 16. Frequency distributions of nighttime radon 222 concentrations from 1830 to 0500 CST.

SUMMARY AND CONCLUSIONS

"Emanating Radium 226" in Particles of Soils

Soil samples collected from a wide area of the United States have been examined for "emanating radium 226" contents. Clay type soils have contained from $(0.342 \text{ to } 0.690) \times 10^{-12}$ grams of "emanating radium 226" per gram of dry soil while sandy soils contain from $(0.147 \text{ to } 0.244) \times 10^{-12}$ grams per gram. The lower activity, as determined by release of radon 222, has been attributed to the fact that sand has a lower surface to volume ratio than clays.

Materials removed from an area where mining for uranium 238 or radium 226 is practiced contained from $(0.45 \text{ to } 14.3) \times 10^{-12}$ grams of "emanating radium 226" per gram of dry soil.

Release of Radon 222 From Surface of Soil Beds

Emanation of radon 222 from the surface of soils was measured in units of 10^{-18} curies cm^{-2} sec^{-1}. Means and 1 standard deviation are reported. In Champaign County, Ill., values ranged from 53 ± 16 to 174 ± 42 units. For 216 measurements over three soil types, which cover 85 percent of the county, the emanation measured was 140 ± 73 units. Measurements at Argonne National Laboratory ranged from 57 ± 5 to 92 ± 30 units, while an emanation of 102 ± 30 units was measured at Socorro, N. Mex. However, at locations where uranium 238 mining is or has been commercially feasible, emanation ranged from 536 to 127,136 units for individual measurements. At Grants, N. Mex., eight measurements were given as $37{,}768 \pm 26{,}862$ units.

Release of Radon 222 From Plants

The ability of leaves of corn (*Zea may L.*) to release radon 222 at their peak of growth and near midday has been investigated. At this time release measured was $(247 \pm 19) \times 10^{-18}$ curies cm^{-2} sec^{-1} for six samples.

Radon 222 in the Atmosphere

A series of measurements of radon 222 concentrations at Argonne National Laboratory provides an indication of the natural variation of that material in the atmosphere. Emanation measurements of

$(57\pm5$ to $92\pm30)\times10^{-18}$ curies cm^{-2} sec^{-1}, while measured for moist soils in both cases, are not greatly different from those measured in Champaign County, Ill., $(140\pm73)\times10^{-18}$ curies cm^{-2} sec^{-1}.

As radon 222 is supplied to the atmosphere at Argonne National Laboratory at a very nominal rate, concentrations observed at the breathing level for humans provide a guide to exposure to this radioacive material. The data in table 5 show that the variation of concentration in pCi/liter ranges from 0.32 to 1.400 at 1 m and 0.018 to 1.170 at 4 m, while the mean for 1 m is 0.301 and the mean for 4 m is 0.259. A comparison of daytime and nighttime means at these elevations also can be made from data in the table.

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